Surface phonons in periodic and Fibonacci Nb/Cu superlattices

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(Received 1 May 1992; revised manuscript received 13 July 1992)

Surface phonons in both periodic and Fibonacci Nb/Cu metallic superlattice systems have been studied by means of the Brillouin light-scattering technique. Measurement of the phase velocity of the surface Rayleigh wave enabled us to compare the elastic properties of these two types of structures with those calculated from the elastic constants of bulk Cu and Nb. The results obtained have shown that the relevant parameter for determining the elastic behavior of quasiperiodic Fibonacci superlattices is the average distance between Nb/Cu interfaces, rather than the quasiperiodicity, which is the governing parameter in x-ray and structural analysis.

I. INTRODUCTION

In recent years the elastic behavior of Nb/Cu periodic superlattices (PSL) has been the subject of both experimental and theoretical investigation. The reason for the interest was the discovery of a marked dependence of the surface Rayleigh wave velocity on the period of PSL.[1–3] This anomaly has been explained using a simple model based on the Murnaghan equation of state, which correlates the elastic anomaly to the observed variation of the lattice constant perpendicular to the interface.[4] Little is known, however, about the elastic response of nonperiodic superlattices, such as quasiperiodic Fibonacci superlattices (QPFSL). These structures are an intermediate state between ordered and disordered materials,[5] so that Brillouin scattering investigation can give a further insight into the role of interface effects on their elastic properties.

The aim of this paper is to compare the results of Brillouin scattering measurements on periodic Nb/Cu superlattices with those obtained in Nb/Cu QPFSL. Measurements of the surface Rayleigh wave (RW) phase velocity are presented and the effect of the interfaces in determining the elastic response of both periodic and Fibonacci superlattices is discussed.

II. CHARACTERISTICS OF THE SPECIMENS

The Nb/Cu superlattices studied were deposited by magnetron sputtering on glass substrates. The thickness of alternate layers was controlled by a quartz resonator located inside the sputtering chamber. Three periodic superlattices have been investigated, all having equithick layers of Nb and Cu. The first is made by 400 bilayers 2.5 nm thick, corresponding to a total thickness of 1 μm. The second consists of 100 bilayers 10.8 nm thick (total thickness 1.08 μm); the third of 50 bilayers 2.3 nm thick (total thickness 1.15 μm). The quasiperiodic Fibonacci superlattice is made by two building blocks A and B, which are superimposed according to the Fibonacci sequence: $S_1=A$, $S_2=AB$, $S_3=ABA,\ldots,S_j=\{S_{j-1}S_{j-2}\}$.[5] In our case the sample is a twelfth generation superlattice; block A consists of 6 nm Cu and 3 nm Nb, so that $d_A=9$ nm; block B consists of 3 nm Cu and 3 nm Nb, so that $d_B=6$ nm. The total thickness is about 1.83 μm and the average lattice parameter, i.e., the quasiperiodicity of this sample, is $p_q=\tau d_A+d_B=20.5$ nm, where $\tau=(1+\sqrt{5})/2$ is the golden mean.

X-ray-diffraction spectra have shown that Nb/Cu superlattices are polycrystalline with a well-defined texture perpendicular to the surface. The orientation of the crystallites is (111) for Cu and (110) for Nb. Due to the random orientation of the crystallites in the surface plane, the structure is elastically isotropic in this plane. The superlattice film can thus be modeled as a hexagonal elastic medium, with the c axis orthogonal to the free surface. Five independent effective elastic constants are required to describe the elastic properties of the structure: referring to a coordinate system with the x_3 axis perpendicular to the surface, these constants are labeled c_{11}, c_{12}, c_{13}, c_{33}, and c_{44}. Starting from the bulk elastic constants of fcc Cu and bcc Nb given in Ref. 6, we have calculated the elastic constants of polycrystalline Cu(111) and Nb(110), taking the Voigt average around the crystallites.[7] These values have been used to calculate the effective elastic constants of periodic superlattices according to the effective modulus model outlined in Ref. 8. With respect to the QPFSL, we have first calculated the effective constants of sublattices A and B, taking into account the fraction of Cu and Nb constituting each of them; the effective elastic constants of the QPFSL have then been obtained simply from those of A and B, provided that a Fibonacci lattice can be regarded as a superposition of the two periodic sublattices A and B with periods τd_A and d_B, respectively.[8] In Table I we show the calculated values of the effective elastic constants of Nb(110), Cu(111), and of Nb/Cu superlattices together with the values of the mass densities and of the surface Rayleigh wave velocity v_R; this latter has been calculated by use of a numerical procedure, based on the theoretical approach of Ref. 9. It can be seen that v_R does not appreciably

TABLE I. Elastic constants of polycrystalline Cu(111), Nb(110), and of periodic and Fibonacci quasiperiodic Nb/Cu superlattices analyzed here. These values are obtained by means of the Voigt average, within the effective-medium approximation. In the last two columns we have reported the mass density of each lattice and the calculated phase velocity of the Rayleigh surface wave v_R.

	c_{11}	c_{13} (GPa)	c_{33}	c_{44}	ρ (g/cm^3)	v_r (m/s)
Nb(110)	220.4	146.0	216.9	42.5	8.570	2053
Cu(111)	220.3	86.8	237.6	40.8	8.960	2079
PSL	216.5	117.7	226.8	41.6	8.765	2086
QPFSL	216.7	110.7	229.3	41.5	8.811	2089

differ for both the constituents Cu(111) and Nb(110) and, consequently, it is almost the same in PSL ($v_R=2086$ m/s) and in QPFSL ($v_R=2089$ m/s). This means that the experimental values of v_R relative to QPFSL and PSL can be directly compared, even if the Cu and Nb content of these two structures is slightly different.

III. EXPERIMENT AND DISCUSSION

Brillouin spectra have been taken in air at room temperature by means of a tandem triple pass Sandercock-type Fabry-Perot interferometer[10,11] having a finesse of about 150 and a contrast ratio greater than 5×10^{10}. The light source is an Ar^+ laser, operating on a single mode of the 514.5-nm line. Both incident and scattered light were polarized in the plane of incidence (*p-p* scattering). Measurements have been taken in the backscattering interaction geometry; in this condition the wave number Q of surface phonons coming into the scattering process is fixed by the conservation of momentum: $Q=2k\sin\theta$, where k is the optical wave number and θ is the incidence angle of light.[11]

Typical Brillouin spectra taken on a periodic and on a quasiperiodic superlattice are shown in Figs. 1(a) and 1(b), respectively. The Rayleigh wave phase velocity v_R has been determined from the measurement of the frequency shift f of the RW peak, according to the relationship

$$v_R=\pi f/k\sin\theta\ .$$

In PSL's we have found a marked increase of this velocity, from 1800 to 2030 m/s, as the period increases from 2.5 to 23.0 nm. This behavior is illustrated in Fig. 2. The horizontal dashed line indicates the expected value of the phase velocity, obtained from the Voigt estimate, on the basis of the effective modulus model, as outlined in the previous paragraph. In the absence of interface anomalies one should find the same value of the phase velocity, whatever the period. Actually, the dispersion of the phase velocity found experimentally indicates the presence of interface anomalies whose effect becomes noticeable when the period of the structure is decreased. In Fig. 2 we have also reported the value of the phase velocity measured on the QPFSL. The experimental point (filled square) is located in correspondence to the quasiperiodicity $P_q=20.5$ nm, which is the relevant parameter with respect to translational order just like the period in the periodic superlattice. We stress that, according to the calculated values of Table I, the Rayleigh wave velocity for the QPFSL should practically coincide with that of the PSL (dashed line in Fig. 2) in the absence of elastic anomalies. It can be seen, however, that the measured value is considerably lower than the one expected on the basis of the PSL experimental data. In order to find the

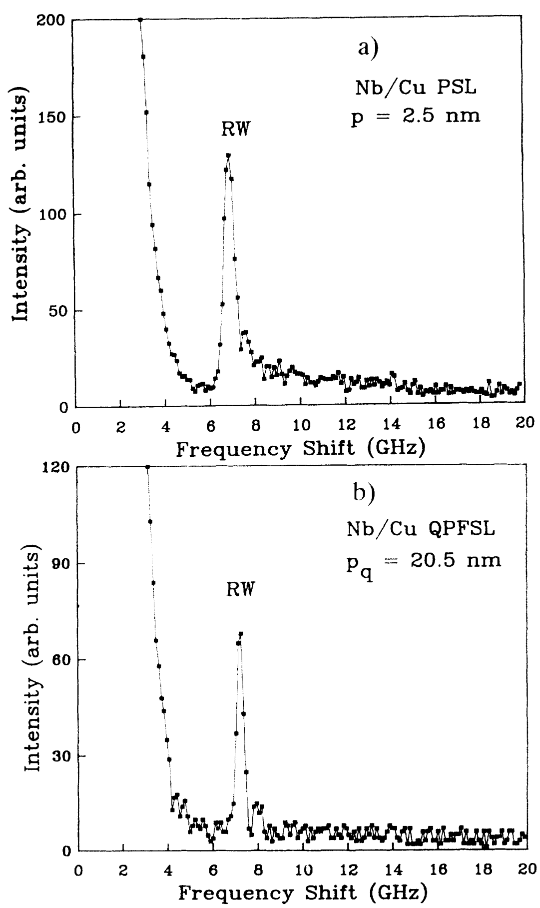

FIG. 1. Brillouin spectra taken on a periodic Nb/Cu superlattice (a), with period $p=2.5$ nm and total thickness 1 μm, and on a Nb/Cu quasiperiodic Fibonacci superlattice (b), with quasiperiodicity $P_q=20.5$ nm and total thickness 1.83 μm; the angle of incidence of light is $\theta=67.5°$. The peak labeled RW is due to the surface Rayleigh wave.

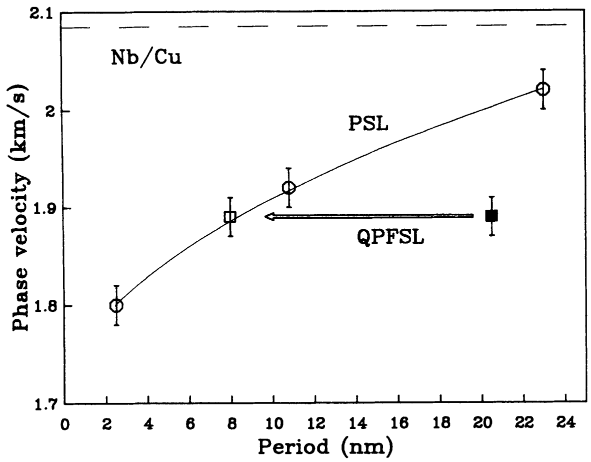

FIG. 2. Measured values of the Rayleigh wave phase velocity as a function of the superlattice period. Open circles refer to PSL's; the solid line is a guide to the eyes. Squares refer to the QPFSL; the arrow indicates how the point relative to the QPFSL must be translated if one assumes the double average distance between Nb/Cu interfaces as the "effective period" of the structure (open square), instead of the quasiperiodicity (filled square). The horizontal dashed line indicates the phase velocity calculated on the basis of the effective modulus model, neglecting interface anomalies.

reason for this discrepancy we must consider the origin of the dependence of the elastic response on the period. Assuming this dependence to be due to interface-induced deformations, we can take the number of Nb/Cu interfaces per unit length, i.e., the average interface density, as the pertinent parameter. In the case of the QPFSL here analyzed, this parameter can be directly determined since we know that sublattice A is made by 6 nm Cu and 3 nm Nb, while B is made by 3 nm of both Cu and Nb. In addition, a high generation Fibonacci lattice can be modeled as a superposition of sublattices A and B, with weighting factors τ and 1, respectively. This means that as an average, block A is τ times more present than block B. Therefore, the average distance between the interfaces in the QPFSL is

$$d_i(\text{QPFSL}) = \frac{\tau d_i(A) + d_i(B)}{\tau+1} = \frac{\tau(3+6)/2+(3+3)/2}{\tau+1} \approx 4 \text{ nm},$$

where $d_1(A)$ and $d_1(B)$ are the interface distances in A and B, respectively. This corresponds to an "effective period" of about 8 nm, i.e., to an average interface density of 0.125 nm^{-1}. From this argument, the experimental point in Fig. 2 should correspond to an effective period of 8 nm, i.e., it must be shifted to the left (open square) as shown by the arrow in the figure. In such a way, the value of the Rayleigh wave phase velocity measured on the Fibonacci superlattice (1890 m/s) appears perfectly consistent with the values measured in periodic superlattices.

IV. CONCLUSION

The phase velocity of the surface Rayleigh wave has been measured on both periodic and Fibonacci quasiperiodic Nb/Cu superlattices. In PSL's our results indicate a strong dependence of the phase velocity on the superlattice period. This dependence is consistent with that observed in previous investigations of Nb/Cu PSL's and can be attributed to interface-induced strains. In Fibonacci superlattices we have found that the phase velocity of surface modes is linked to the average Nb/Cu interface density rather than to the quasiperiodicity, which is the relevant parameter in x-ray and other structure-sensitive techniques. This result is a further step toward the comprehension of the origin of elastic anomalies in artificially created metallic multilayers.

[1] A. Kueny, M. Grimsditch, K. Miyano, I. Banerjee, C. M. Falco, and I. K. Schuller, Phys. Rev. Lett. **48**, 166 (1982).
[2] J. A. Bell, W. R. Bennett, R. Zanoni, G. I. Stegeman, C. M. Falco, and C. T. Seaton, Solid State Commun. **64**, 1339 (1987).
[3] I. K. Schuller and M. Grimsditch, J. Vac. Sci. Technol. B **4**, 1444 (1986).
[4] M. H. Grimsditch, in *Light Scattering in Solids V*, edited by M. Cardona and G. Guntherodt (Springer-Verlag, Berlin, 1989), p. 285.
[5] R. Merlin, IEEE J. Quantum Electron. **24**, 1791 (1988).
[6] R. F. S. Hearmon, in *Landolt-Börnstein Numerical Data and Functional Relationships in Science and Technology, New Series, Vol. 11*, edited by K. H. Hellwege and A. M. Hellwege (Springer-Verlag, Berlin, 1979), p. 11
[7] M. J. P. Musgrave, *Crystal Acoustics* (Holden-Day, San Francisco, 1970), p. 177.
[8] E. Akcakaya, G. W. Farnell, and E. L. Adler, J. Appl. Phys. **68**, 1009 (1990).
[9] G. W. Farnell and E. L. Adler, in *Physical Acoustics*, edited by W. P. Mason and R. M. Thurston (Academic, New York, 1972), Vol. 9, p. 35.
[10] G. Carlotti, D. Fioretto, L. Palmieri, G. Socino, L. Verdini, and E. Verona, IEEE Trans. Ultrason. Ferroelectr. Freq. Contr. **38**, 56 (1991).
[11] F. Nizzoli and J. R. Sandercock, in *Dynamical Properties of Solids*, edited by G. K. Horton and A. A. Maradudin (North-Holland, Amsterdam, 1990), Vol. 6, p. 307.